Polarization dependence of spin excitations in $BaCu_2Si_2O_7$.

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The polarization dependence of magnetic excitations in the quasi one-dimensional antiferromagnet $BaCu_2Si_2O_7$ is studied as a function of momentum and energy transfer. The results of inelastic neutron scattering measurements are directly compared to semi-analytical calculations based on the chain-Mean Field and Random Phase approximations. A quantitative agreement between theoretically calculated and experimentally measured dynamic structure factors of transverse spin fluctuations is obtained. In contrast, substantial discrepancies are found for longitudinal polarization. This behavior is attributed to intrinsic limitations of the RPA that ignores correlation effects.

I. INTRODUCTION

Excitations in weakly ordered quasi-one-dimensional (quasi-1D) antiferromagnets (AFs) are a topic of considerable current interest in the field of quantum magnetism. Particularly intriguing is the problem of the so-called *longitudinal mode* (LM), a magnon excitation polarized *parallel* to the direction of ordered moment. The discovery of a coherent LM in $KCuF_3$ (Refs. 1,2) confirmed previous theoretical predictions,[3] based on the chain-Mean Field[4] (chain-MF) and Random Phase Approximation (RPA) theories.[3,5] Currently chain-MF/RPA indeed appears to be the most versatile analytical framework for treating weakly-coupled quantum spin chains. However, the $KCuF_3$ experiments also highlighted certain limitations of this approach. In particular, the chain-MF/RPA can not, by its very definition, account for the experimentally observed finite lifetime (broadening) of the LM.

In a recent short paper[6] we reported polarization-sensitive neutron scattering measurements of the dynamic spin structure factor in another model quasi-1D antiferromagnet, namely $BaCu_2Si_2O_7$. This $S = 1/2$ system has much weaker inter-chain interactions and low-temperature ordered moment than $KCuF_3$. Preliminary results indicated that, unlike in $KCuF_3$, in $BaCu_2Si_2O_7$ there is *no well-defined longitudinal mode*. Instead, the longitudinal spectrum is best described as a single broad asymmetric continuum feature. This stark discrepancy with the predictions of the chain-MF/RPA model came as surprise. Indeed, for the *transverse*-polarized spectrum of $BaCu_2Si_2O_7$, earlier neutron scattering work confirmed excellent agreement with chain-MF/RPA theory, at least as far as excitation energies were concerned.[7,8] The apparent paradox is not fully resolved to date. This is in part due to that only very limited data are available for longitudinal-polarized excitations. Even for the transverse-polarized spectrum, the existing wealth of high-resolution neutron data could not be *quantitatively* compared to theoretical predictions, for lack of calculations based on the specific geometry of inter-chain interactions in $BaCu_2Si_2O_7$. The present work addresses both these issues and involves a detailed experimental and theoretical study of the polarization dependence of magnetic excitations in this compound. First, we further exploit the technique of polarization analysis described in Ref. 6 to investigate the wave vector dependence of longitudinal excitations. We then perform chain-MF/RPA calculations of the dynamic structure factor for the exchange topology and constants of $BaCu_2Si_2O_7$. This enables us to perform a direct *quantitative* comparison between theory and experiment for both energies and *intensities* of the coherent and diffuse components of the dynamic spin correlation functions.

Magnetic interactions in $BaCu_2Si_2O_7$ have been previously thoroughly studied using bulk methods,[9,10] neutron diffraction,[9,11] and inelastic neutron scattering.[8,9,12,13]

The silicate $BaCu_2Si_2O_7$crystallizes in an orthorhombic structure (space group $Pnma$, $a = 6.862$ Å, $b = 13.178$ Å, $c = 6.897$ Å) with slightly zigzag AF $S = 1/2$ chains of Cu^{2+} ions running along the c axis. The in-

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FIG. 1: Typical constant-E scans (a,b) and constant-Q scans (c) measured in $BaCu_2Si_2O_7$ in magnetic fields $H = 1.5$ T (open circles) and $H = 2.2$ T (solid circles) applied along the crystallographic c-axis. The dashed lines in (a) and (b) represent the background obtained by linear interpolation between intensities measured at $l = 0.8$ and $l = 1.2$. In (c) the dashed line is the background scan measured at $\mathbf{q} = (0, -0.5, 1.2)$.

respectively.[10,11] The transition involves a re-orientation of the ordered staggered magnetization in the system.[11] As explained in Ref. 6, this leads to a drastic change in the polarization-dependent part of the scattering cross section for unpolarized neutrons. The effect on the scattering intensity from longitudinal (parallel to the ordered moment) and transverse (perpendicular to the ordered moment) spin fluctuations is different, which allows us to separate the two components. In general, the measured intensity can be expanded as:

$$\begin{aligned} I(\mathbf{q},\omega) \;&\propto\; S^{\perp}(\mathbf{q},\omega)(1+\cos^2\alpha_{\mathbf{q}}) \\ &+\; S^{\parallel}(\mathbf{q},\omega)\sin^2\alpha_{\mathbf{q}} + \mathcal{B}(\mathbf{q},\omega). \end{aligned} \tag{1}$$

In this equation $S^{\perp}(\mathbf{q},\omega)$ and $S^{\parallel}(\mathbf{q},\omega)$ are the magnetic dynamic structure factors for transverse and longitudinal polarizations, respectively. The wave vector dependent angle $\alpha_{\mathbf{q}}$ is measured between the momentum transfer $\mathbf{q}$ and the direction of ordered moment. The orientations of the latter was previously determined using neu-

FIG. 3: Transverse (a) and longitudinal (b) components of a constant-Q scan measured in $BaCu_2Si_2O_7$ at $\mathbf{q} = (0, -0.5, 1)$. Lines and the plot shown in (c) are as in Fig. 2.

detail in Ref. 6.

III. EXPERIMENTAL RESULTS

Typical raw data sets measured in constant-Q and constant-E modes at $T = 1.5$ K are shown in Fig. 1. At energies in excess of about 2Δ the scattering is practically unaffected by the phase transition (Fig. 1b). The contrast in inelastic intensity measured at two different field values is most apparent at energy transfers of about Δ(Fig. 1a and c). Separating the longitudinal and transverse contributions as described in the previous section yields the constant-Q scans shown in Figs. 2–4. The evolution of the instrumental FWHM resolution ellipsoid in the course of each scan is shown in the right part of each figure. Typical constant-E data are shown in Fig. 5. A contour and false color plot based on a series of 10 such scans taken with 1 meV energy step is shown in Fig. 6.

Certain important features of the measured transverse and longitudinal dynamic structure factors can be identified even without a quantitative data analysis. An important experimental observation is that longitudinal excitations show a steep dispersion along the chains. As can be seen in Fig. 6, the corresponding spin velocity is the same as for transverse-polarized spin waves. Furthermore, at high energy transfers (above ≈ 7 meV) the scattering is almost *polarization-independent* to within experimental accuracy and resolution (Figs. 2–5). Such behavior is consistent with our general expectation that inter-chain interactions become almost irrelevant at energies well above the gap energy Δ. The dynamic structure factor in this regime is as in isolated chains, and is therefore almost isotropic.

At smaller energy transfers the structure factors for longitudinal and transverse polarizations are noticeably different. As observed in previous detailed studies,[8] transverse-polarization constant-Q scans are character-

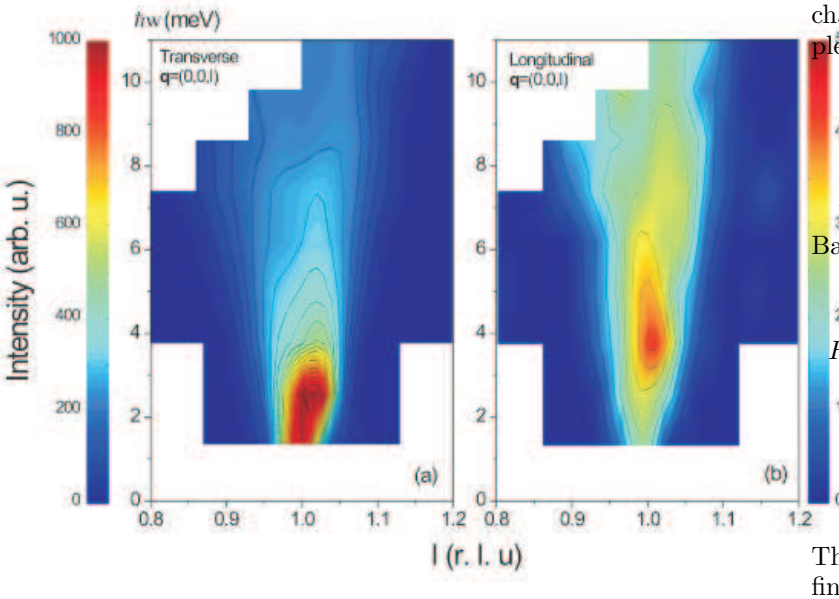

FIG. 6: Contour and false-color plot of the transverse-polarized (left) and longitudinal-polarized (right) inelastic scattering measured in $BaCu_2Si_2O_7$ near the 1D AF zone-center $(0, 0, 1)$.

ized by a sharp spin wave peak, whose position and intensity is strongly dependent on momentum transfer $\mathbf{q}_\perp$ in the direction perpendicular to the spin chains. The effect of this pronounced dispersion can be seen in Figs. 2a–4a. In contrast, longitudinal-polarized scans lack the sharp component and are *almost independent of* $\mathbf{q}_\perp$ (Figs. 2b–4b). Such behavior is reminiscent of that for the transverse-polarized *continuum* that also shows very little variation with $\mathbf{q}_\perp$.[8]

IV. THEORY

Before discussing the quantitative analysis of the experimental data we shall describe the application of the chain-MF/RPA approach to the problem of weakly coupled chains in $BaCu_2Si_2O_7$.

A. Hamiltonian and definitions

Following Refs. 8,13 the spin Hamiltonian for $BaCu_2Si_2O_7$ is written as:

$$\begin{aligned} H &= H_{\rm chains} + H' \,, \\ H_{\rm chains} &= J \sum_{i,j,n} \mathbf{S}_{i,j,n} \cdot \mathbf{S}_{i,j,n+1} \,, \\ H' &= \sum_{i,j,n} J_x \, \mathbf{S}_{i,j,n} \cdot \mathbf{S}_{i+1,j,n} + J_y \, \mathbf{S}_{i,j,n} \cdot \mathbf{S}_{i,j+1,n} \\ &\quad + J_3 \, \mathbf{S}_{i,j,n} \cdot (\mathbf{S}_{i+1,j+1,n} + \mathbf{S}_{i+1,j-1,n}) \,. \end{aligned} \quad (2)$$

The Fourier transform of the inter-chain coupling is defined as

$$\begin{aligned} J'(\mathbf{q}) &= J_x \cos(q_x) + J_y \cos(q_y) \\ &\quad + J_3 \left[\cos(q_x + q_y) + \cos(q_x - q_y)\right] . \end{aligned} \quad (3)$$

$$h \;=\; 2(J_x - J_y - 2J_3)\; m_0 \equiv J' m_0. \qquad (7)$$

The Hamiltonian (7) describes an ensemble of *uncoupled* spin-$\frac{1}{2}$ Heisenberg chains in a staggered magnetic field

$$H_{\rm 1d} = \sum_n J\widetilde{\mathbf{S}}_n \cdot \widetilde{\mathbf{S}}_n + h(-1)^n\; \widetilde{S}^z_{n+1}\;. \qquad (8)$$

The next step is to find a solution for an isolated chain in an external field h. Since in the limit of weak inter-chain coupling the latter is expected to be small compared to J, it is possible to determine dynamical correlation functions at low energies $\hbar\omega \ll J$ by means of field theory methods. A standard bosonization analysis gives the following scaling limit of (8):

$$\mathcal{H}_{\rm 1d} \;=\; \int dx \left[\frac{v}{2}\left(\partial_x\phi\right)^2 + \frac{1}{2v}\left(\partial_t\phi\right)^2 + Ch\cos(\sqrt{2\pi}\phi)\right]. \qquad (9)$$

In this formula $v = \pi J a_0/2$ is the spin velocity of the spin-1/2 Heisenberg chain[22] and C is a non-universal constant that was calculated in Ref. 14. The model (9) is known as the quantum Sine Gordon model (SGM) and is exactly solvable. The spectrum is formed by scattering states of four particles, called soliton s, antisoliton $\bar{s}$, breather B_1 and breather B_2. The breathers themselves are soliton-antisoliton bound states. All four particles have gapped relativistic dispersion relations:[23]

$$E_\alpha \;=\; \Delta\cosh\theta\;, \quad P_\alpha = \frac{\Delta}{v}\sinh\theta\;,\; \alpha = s, \bar{s}, B_1\;,$$

$$E_{B_2} \;=\; \sqrt{3}\Delta\cosh\theta\;, \quad P_{B_2} = \frac{\sqrt{3}\Delta}{v}\sinh\theta. \qquad (10)$$

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FIG. 8: Comparison of simulated scans across transverse-polarized continuum at $\mathbf{q} = (0, 0, 1)$ based on the exact chain-MF/RPA result (solid line) and the empirical TMA fitting function that was used to analyze the neutron scattering data (dotted line). Given the effects of experimental resolution that were taken into account in these simulations, the two curves are virtually identical.

Here $\omega_{\perp}^2(\mathbf{q})$ is the spin wave dispersion relation given by:

$$\omega_{\perp}^2(\mathbf{q}) = \frac{\pi^2}{4} J^2 \sin^2(\pi l) + \frac{\Delta^2}{|J'|} \left[|J'| + 2J'(\mathbf{q}) \right], \qquad (24)$$

where $J'(\mathbf{q})$ is defined by Eq. 3.

The second component of the fit function for transverse excitations approximates the continuum. We have previously found that, at least for wave vectors on the $(0, k, 1)$ reciprocal-space rod, continuum scattering can be very well approximated by the "truncated Müller ansatz" (TMA) function:[24]

$$\begin{aligned} S_{\mathrm{c}}^{\perp}(\mathbf{q}, \omega) \;&=\; \frac{\alpha A}{2} \frac{[1 - \cos(\pi l)]}{\sqrt{\omega^2 - \frac{\pi^2}{4} J^2 \sin^2(q_{\parallel})}} \times \\ &\times\; \theta \left(\omega^2 - \Delta_{c,\perp}^2 - \frac{\pi^2}{4} J^2 \sin^2(\pi l) \right) \end{aligned} \qquad (25)$$

The TMA is plotted in a thin dotted lined in Figs. 7a–c for a direct comparison to our chain-MF/RPA result. Conveniently, given the experimental resolution width, the two functional forms are almost indistinguishable. This fact is illustrated in Fig. 8 that shows the transverse continuum obtained in the actual chain-MF/RPA calculation for $\mathbf{q} = (0, 0, 1)$ (solid line), along with the form 25 (dotted line), both profiles being numerically convoluted with the resolution function of the instrument. Resolution effects taken into account, an almost perfect match between the chain-MF/RPA calculation for $BaCu_2Si_2O_7$ and Eqs. (23) and (25) can be obtained for the entire range of energy and momentum transfers covered in our experiments by choosing $\Delta_{c,\perp} = 5.0$ meV and $\alpha = 0.17$ meV^{-1}.

Just like the transverse-polarized part, the fit function for longitudinal scattering is composed of a single-mode and a continuum components. The dispersion relation and dynamic structure factor for the single-mode contribution are written as:

$$S^{\parallel}_{\rm SM}(\mathbf{q},\omega) = \frac{\gamma}{2} A \frac{[1-\cos(\pi l)]}{2\omega_{\parallel}(\mathbf{q})} \frac{\Gamma/\pi}{\left[\omega-\omega_{\parallel}(\mathbf{q})\right]^2+\Gamma^2} \quad (26)$$

$$\omega^2_{\parallel}(\mathbf{q}) = \frac{\pi^2}{4} J^2 \sin^2(\pi l) + \Delta^2_{\parallel} + \frac{\Delta^2 J'(\mathbf{q})}{|J'|} \quad (27)$$

These equations are a generalization of Eqs. (12) and (13) in Ref. 8, that allow for a damping of the longitudinal mode. The adjustable parameter $\Delta_{\parallel}$ is the the energy of the longitudinal mode at the RPA "magic" point. The coefficient γ is an adjustable parameter that determines the intensity ratio of longitudinal and transverse excitations, while A is an overall intensity prefactor used for both polarizations (see Eqs. (4), (5), and (10) in Ref. 8). In Eq. (26) the δ-function is replaced (for positive energy transfers) by a Lorentzian profile with a half-width at half height of Γ.

The longitudinal-polarized excitation continuum was modelled using the same truncated Müller-ansatz cross section function as previously done for the transverse case:

$$S^{\parallel}_{\rm c}(\mathbf{q},\omega) = \frac{\beta A}{2} \frac{[1-\cos(\pi l)]}{\sqrt{\omega^2-\frac{\pi^2}{4}J^2\sin^2(q_{\parallel})}} \times \times \theta\left(\omega^2-\Delta^2_{c,\parallel}-\frac{\pi^2}{4}J^2\sin^2(\pi l)\right) \quad (28)$$

Note that, unlike in Ref. 8, we use separate relative intensity prefactors and (pseudo)gap energies for the transverse and longitudinal continua. By choosing $\Delta_{c,\parallel} = \Delta_{c,\perp}$ and $\beta = \alpha$ one can accurately reproduce the chain-MF/RPA result for $BaCu_2Si_2O_7$ to within resolution effects in the energy and momentum transfer range covered in the present study.

B. Transverse polarization

As a first step in the quantitative data analysis, the two-component model cross section for transverse polarization was numerically convoluted with the calculated spectrometer resolution function and fit to the transverse components of all scans measured in this work (429 total scan points). The relevant parameters of the model, including the mass gap $\Delta = 2.51(2)$ meV, the inter-chain exchange constants $J_x = -0.460(7)$ meV, $J_y = 0.200(6)$ meV, $2J_3 = 0.152(7)$ meV, the in-chain exchange parameter $J = 24.1$ meV, the continuum gap $\Delta_{c,\perp} = 4.8(2)$ meV, and the ratio $\alpha = 0.20(3)$ meV^{-1} of single-mode and continuum intensities, were determined previously with very good accuracy.[8,13] In analyzing the present data, only the overall scaling factor was treated as an adjustable parameter. A good ($\chi^2 = 2.7$) 1-parameter *global* fit to all the measured scans was obtained (heavy solid lines in Figs. 2a–4a and Figs. 5a–g. The hatched and greyed areas represent the continuum and single-mode components, respectively.

As mentioned in the previous section, our chain-MF/RPA theoretical result for $BaCu_2Si_2O_7$ corresponds to fitting function parameters $\Delta_{c,\perp} = 2\Delta = 5.0$ meV and $\alpha = 0.17$ meV^{-1}, which is in a remarkably good agreement with previous and current experiments. We conclude that for transverse polarization the chain-MF/RPA not only predicts the correct spin wave dispersion relation and continuum gap energy, but provides an accurate estimate for the *intensity* of the lower-energy part of the continuum.

C. Longitudinal polarization

The agreement with theory is not nearly as good in the longitudinal polarization channel. In the chain-MF/RPA the LM is infinitely sharp and corresponds to $\Gamma \to 0$ in Eq. (26). The LM's energy and intensity are given by $\Delta_{\parallel} = \sqrt{3}\Delta$, and $\gamma \approx 0.49$. Our chain-MF/RPA calculation for the longitudinal continuum in $BaCu_2Si_2O_7$ is very well approximated by Eq. (28) with $\Delta_{c,\perp} = \Delta_{c,\parallel}$ and $\beta = \alpha$. Using these values in the model cross section convoluted with the resolution function of the spectrometer, we can simulate the measured scans as expected in the chain-MF/RPA model. These simulations are shown in solid lines in Figs. 2b–4b. The dark greyed area represents the longitudinal mode, and the hatched area is the continuum contribution. It is clear that at all values of $\mathbf{q}_{\perp}$ the model fails to reproduce the observed longitudinal spectrum. The discrepancy is greatest at energy transfers *below* 2Δ, where the chain-MF/RPA model predicts no scattering except that by the LM. At higher energy transfers the agreement between theory and experiment becomes progressively better.

Of course, much better fits to the experimental data can be obtained if the central energy $\Delta_{\parallel}$, intensity prefactor γ and intrinsic energy width Γ of the longitudinal mode are allowed to vary. The result of fitting this "damped LM" model globally to the entire data set for longitudinal polarization (358 data) is shown in Figs. 2b–4b in a dotted line, and corresponds to $\chi^2 = 1.5$. The fit yields $\Delta_{\parallel} = 2.1(1)$ meV, $\gamma = 1.2(2)$ and $\Gamma = 1.5(2)$ meV. This analysis confirms the main conclusion of the preliminary study of Ref. 6: to adequately describe the longitudinal scattering in $BaCu_2Si_2O_7$ in terms of a "longitudinal mode" one has to assume a substantial intrinsic width, comparable to the mode's central energy and to its separation from the continuum threshold. The "longitudinal mode" can therefore be no longer considered a separate feature, as it is merged with the strong continuum at higher energy transfers. The *energy separation* of single-mode and continuum excitations previously observed for transverse polarization is *absent in the longitudinal chan-*

nel. It is important to emphasize that the mismatch between theory and experiment involves more than simply a broadening of the LM. Experimentally one observed considerably more scattering below 2Δ energy transfer than the LM could provide in the chain-MF/RPA model. As a result, the refined value of γ is almost 4 times larger than expected, and the "LM" is almost equal in intensity to a transverse spin wave.

The measured data can, in fact, be reproduced without including a single-mode longitudinal component in the cross section. This "continuum-only" model corresponds to $\gamma = 0$, while $\Delta_{c,\|}$ and $\alpha_\|$ are the adjustable parameters. Rather good global fits to 249 data points at $k = 0$ are obtained with $\Delta_{c,\|} = 2.0(1)$ meV, $\alpha_\| = 0.22(0.01)$ and $\chi^2 = 1.16$. Scan simulations based on these parameter values are plotted in heavy solid lines in Fig. 5b,d,f, and h, and in a dash-dot line in Fig. 2b. The parameter $\Delta_{c,\|}$ was fit separately for the constant-Q scans at $k = -0.5$ and $k = -1$, yielding $\Delta_{c,\|} = 1.8(1)$ meV and $\Delta_{c,\|} = 1.5(1)$ meV, respectively. The results are shown in dash-dot lines in Figs. 3b and 4b. The variation of $\Delta_{c,\|}$ as a function of $\mathbf{q}_\perp$ corresponds to the dispersion of the longitudinal mode built into the "damped LM" model.

VI. CONCLUDING REMARKS

Based on the neutron scattering results we can now give phenomenological description of the longitudinal excitations in weakly interacting quantum spin chains. There is no sharp longitudinal mode, but a broad asymmetric peak that is inseparable from the continuum at higher frequencies. This feature is practically independent of $\mathbf{q}_\perp$, but has a steep dispersion along the chain axis. The scattering starts at energies well below 2Δ, and its intensity at low energies is considerably greater than predicted by the chain-MF/RPA.

It appears that the established chain-MF/RPA model is at the same time remarkably good in predicting the transverse correlations of weakly-coupled chains, and sourly inadequate as far as longitudinal fluctuations are concerned. Admittedly, one can never entirely dismiss the possibility that the disagreement between theory and experiment in the latter case may, in fact, be due to some intrinsic flaw in the unconventional technique that we used for polarization analysis. However, having repeatedly scrutinized the measurement procedure, we were unable to identify any potential sources of systematic error that could account for the observed discrepancies with theoretical calculations. We thus conclude that the discrepancies stem from limitations of the theoretical method itself. Among the assumptions and approximations associated with the chain-MF/RPA approach, the most likely source of errors is the uncontrolled discarding of the self-energies in the RPA. The RPA, by definition, acts on bare (purely 1D) dynamic susceptibilities at *particular wave vectors*. It excludes interactions between particles, such as processes that involve a decay of a particle with momentum $\mathbf{q}$ into a pair of particles with momenta $\mathbf{q}_1 + \mathbf{q}_2 = \mathbf{q}$. The contributions of such processes to the susceptibility involve 1D correlation functions of three or more spin operators. For spin chains that are *intrinsically* gapped such processes are expected to be suppressed, in which case the RPA will be fully justified. We can expect the RPA to be an almost perfect description of weakly coupled ladders or Haldane spin chains. For weakly coupled $S = 1/2$ chains however the mean-field gap Δ is itself determined by J'. As a result, the transverse spectrum in RPA is gapless, regardless of J'/J. Hence a longitudinal excitation can always decay into a pair of transverse-polarized spin waves. The RPA fails by excluding this effect. Comparing the results of the present study to the ones reported in Ref.1 for $KCuF_3$, the question arises why there is a longitudinal mode, albeit damped, in the latter material but not in $BaCu_2Si_2O_7$. The main difference between the two materials is the strength of the interchain coupling: in $BaCu_2Si_2O_7$ the ratio of the bandwidths perpendicular to the chains and along the chains is $2\Delta/\pi J \simeq 0.066$, whereas it is approximately 0.2 for $KCuF_3$. This may suggest that a sufficiently strong dispersion perpendicular to the chains is necessary in order to stabilize a damped longitudinal mode. It would be interesting to investigate this issue by determining the damping of the longitudinal mode in MF/RPA.

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[21] In other words, only the first term in Eq. (3) of Ref. 8 is of any importance in the studied **q**-range.
[22] a_0 is the period of the spin chains that for $BaCu_2Si_2O_7$ is equal to $c/2$.
[23] In Ref. 3 the mass gap Δ is denoted as M.
[24] In Ref. 8 there is a a factor of 2 mistake in the quoted value of α. Note also that α is measured in reciprocal energy units.